

THERMOPHYSICAL PROPERTIES OF Te-BASED II-VI SEMICONDUCTORS: REDUCED ALGORITHMS FOR THERMAL DIFFUSIVITY DETERMINATION

R. Michael Banish^{1*}, Segolene Brantschen¹, Timothée L. Pourpoint¹, Francis Wessling¹ and,
Robert F. Sekerka²

¹University of Alabama in Huntsville, ²Carnegie Mellon University

Abstract

This paper presents methodologies for measuring the thermal diffusivity using the difference between temperatures measured at two, essentially independent, locations. A heat pulse is applied for an arbitrary time to one region of the sample; either the inner core or the outer wall. Temperature changes are then monitored versus time. The thermal diffusivity is calculated from the temperature difference versus time. No initial conditions are used directly in the final results.

I. Introduction

The determination of the thermal diffusivity κ (or conductivity k) typically requires precise knowledge of the applied parameters. Such parameters include the time at which a heat pulse was applied, the amount of energy added or the initial temperature increase. The diffusivity is then obtained by matching the experimentally measured profile to the output of an analytical/numerical model. The resulting diffusivity is sensitive to values of these applied parameters.

In most circumstances, measuring thermal diffusivities in liquids is complicated by convection that occurs in the sample due to the applied temperature gradient. See [1,2] for a discussion of convective flows due to density gradients in a gravitational field. In metals, which have relatively high values of κ , this convective contribution will typically be less than a percent. However, for low thermal conductivity liquids, e.g., the II-VI semiconductors, convective heat transport may be important relative to heat conduction. Earlier studies have confirmed the advantage of making such measurements in a low-gravity environment [3]. We plan to measure the thermal diffusivity of molten II-VI semiconductors, for which convective contamination in the data is expected. Therefore, we are looking for a methodology that is efficient and simple for conducting measurements of thermal diffusivities in orbiting spacecraft.

In general, this paper presents a methodology for measuring the thermal diffusivity using the difference between the temperatures measured at two locations in cylindrical samples. We have developed algorithms that allow us to use either heating of the center portion of the sample or the samples edge. The final form of the equation used to calculate the thermal diffusivity is the same for either heating scheme. For both heating geometries the heat pulse is applied for an arbitrary. Temperature changes are then monitored versus time and the thermal diffusivity is calculated from the temperature differences only versus time. No initial conditions are used directly in the final result.

Keywords: thermal diffusivity, thermal conductivity, flight

*Corresponding author. e-mail: banishm@email.uah.edu

In section II-A of this paper, we describe the mathematical formulation of the problem for central region heating. In section II-B we briefly describe the mathematical formulation of the problem for edge heating. Then by means of numerical simulations we examine the effect of variations in the heated region area and measurement locations (Sec. II-C and D). Finally, in Sec. II-E we examine our analytical solution to explain why these effects make such a small difference in the results, considering the stringent conditions that we started with. Section III covers our experimental set-up and results for fine-grained graphite and boron nitride.

II. Temperature In A Cylindrical Rod: The General Problem

II-A Mathematical Formulation for Central Region Heating

We consider a perfectly insulated circular cylinder under conditions for which the temperature depends only on the cylindrical radius r . The initial temperature distribution is $T(r, 0) = f(r)$ given by

$$f(r) = \begin{cases} T_0 & \text{for } 0 < r/a < p \\ 0 & \text{for } p < r/a < 1 \end{cases} \quad (1)$$

where $0 < p < 1$; a is the cylinder radius, and T_0 is a constant, see Fig. 1. For time $t > 0$, radial heat transfer begins.

The evolution of the temperature $T(r, t)$ is then given [4] by the expression

$$T(r, t) = \frac{2}{a^2} \left(\int_0^a r f(r) dr + \sum_{n=1}^{\infty} \exp(-\kappa \alpha_n^2 t) \frac{J_0(r \alpha_n)}{J_0^2(a \alpha_n)} \int_0^a r f(r) J_0(\alpha_n r) dr \right) \quad (2)$$

where $\mu_n = a \alpha_n$ is the n^{th} positive root of the Bessel function of the first kind of order 1, i.e., $J_1(\mu_n) = 0$. Using the initial conditions defined above, and after several substitutions the integrals in Eq. (2) become

$$T(r, t) = T_0 p^2 + T_0 \sum_{n=1}^{\infty} \exp\left(\frac{-\kappa \mu_n^2 t}{a^2}\right) \frac{2p}{\mu_n} \frac{J_0(\mu_n r/a)}{J_0^2(\mu_n)} J_1(\mu_n p) \quad (3)$$

We shall proceed to simplify the infinite sum in Eq. (3) by eliminating the $n = 2$ and $n = 3$ terms. Since the terms for $n > 3$ decay rapidly with time, we will be left after a short transient with $n = 1$ term, ***a tractable expression that can be used to analyze experimental data.*** Setting p , the ratio of the heated to the sample radius, equal to the special value $\mu_1/\mu_2 = 0.5436$, we can eliminate the $n = 2$ term of the series because

$$J_1(\mu_2 p) = J_1(\mu_1) = 0. \quad (4)$$

The $n = 3$ term can be set to zero by choosing the temperature measurement locations r so that $\mu_3 r/a = v_n$ corresponds to the first three zeros of J_0 , i.e., $J_0(v_n) = 0$, which are 2.4048, 5.5201 and 8.6537, respectively. With $\mu_3 = 10.17347$, this results in three special values of r given by $r_1/a = v_1/\mu_3 = 0.2364$, $r_2/a = v_2/\mu_3 = 0.5426$, and $v_3/\mu_3 = v_3/\mu_3 = 0.8506$. The next zero of J_0 is at $v_4 = 11.792$ and would result in a value $r_4/a > 1$, outside the sample.

The geometry of the heated area and the three special values of r (temperature measurement locations for the experiments) are shown in Fig. 1. With the $n = 2$ and $n = 3$ terms set to zero by means of the choices adopted above, the temperature at each r_i is,

$$T(r_i, t) = T_0 p^2 + T_0 \frac{2p}{\mu_1} \frac{J_0(\mu_1 r_i/a)}{J_0^2(\mu_1)} J_1(\mu_1 p) \exp\left(\frac{-\kappa \mu_1^2 t}{a^2}\right) + h.o.t. \quad (5)$$

where h.o.t. stands for higher order terms having $n > 3$.

Taking the difference between two locations, for example between r_1 and r_3 ,

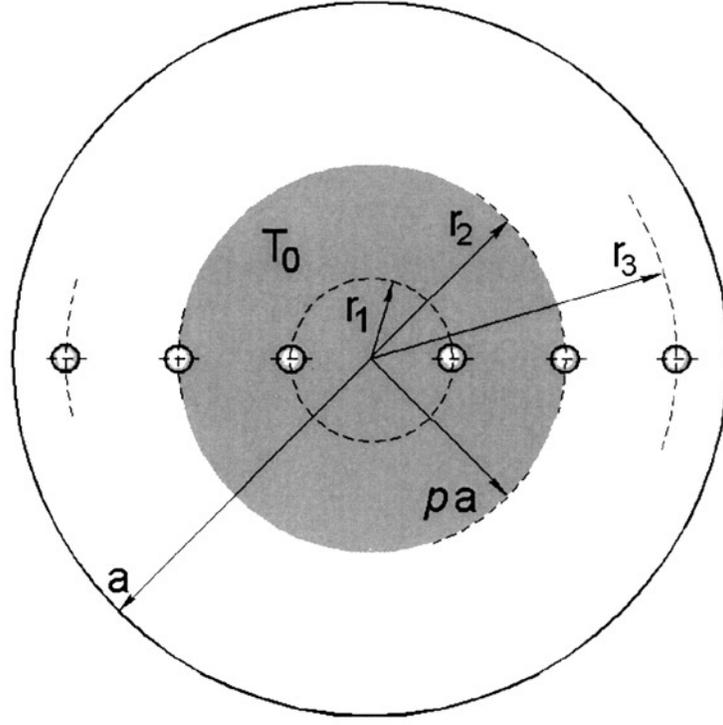


Fig. 1 Plan view of the sample showing the positions of the heated region and temperature measurement locations for central heated region.

$$T(r_1, t) - T(r_3, t) = T_0 \frac{2p}{\mu_1} \frac{J_1(\mu_1 p)}{J_0^2(\mu_1)} \left(J_0\left(\mu_1 \frac{r_1}{a}\right) - J_0\left(\mu_1 \frac{r_3}{a}\right) \right) \exp\left(\frac{-\kappa \mu_1^2 t}{a^2}\right) + \Delta(h.o.t.) \quad (6)$$

It can be shown that the difference between the higher order terms in Eq. (6) is less than 1% of the $n = 1$ term for $\kappa t/a^2 > 0.015$. This corresponds to a time greater than 3.7 s for $a = 5$ cm and $\kappa = 0.1$ cm²/s. The effects of this condition will be discussed in section II-C. Neglecting such high order terms, and taking the natural logarithms of the differences $\Delta T_{ij} = T(r_i, t) - T(r_j, t)$, we obtain

$$\ln(\Delta T_{ij}) = \ln(\beta_{ij}) - \left(\frac{\kappa \mu_1^2}{a^2}\right) t \quad (7)$$

where

$$\beta_{ij} = T_0 \frac{2p}{\mu_1} \frac{J_1(\mu_1 p)}{J_0^2(\mu_1)} \left(J_0\left(\mu_1 \frac{r_i}{a}\right) - J_0\left(\mu_1 \frac{r_j}{a}\right) \right). \quad (8)$$

Details of all of the initial conditions, including the initial applied temperature T_0 , are contained in β_{ij} . Therefore, the slope of a plot of $\ln(\Delta T_{ij})$ versus t is independent of the details of the initial conditions and is proportional to the value of κ , with a known proportionality constant.

The values of thermal diffusivity are obtained from the slopes of the curves of $\ln(\Delta T_{ij})$ as a function of time, as shown in Fig. 2. Note that the resulting diffusivity depends only on the slope of the curve, i.e., we have met the requirement that no knowledge of initial conditions are necessary for the final analysis.

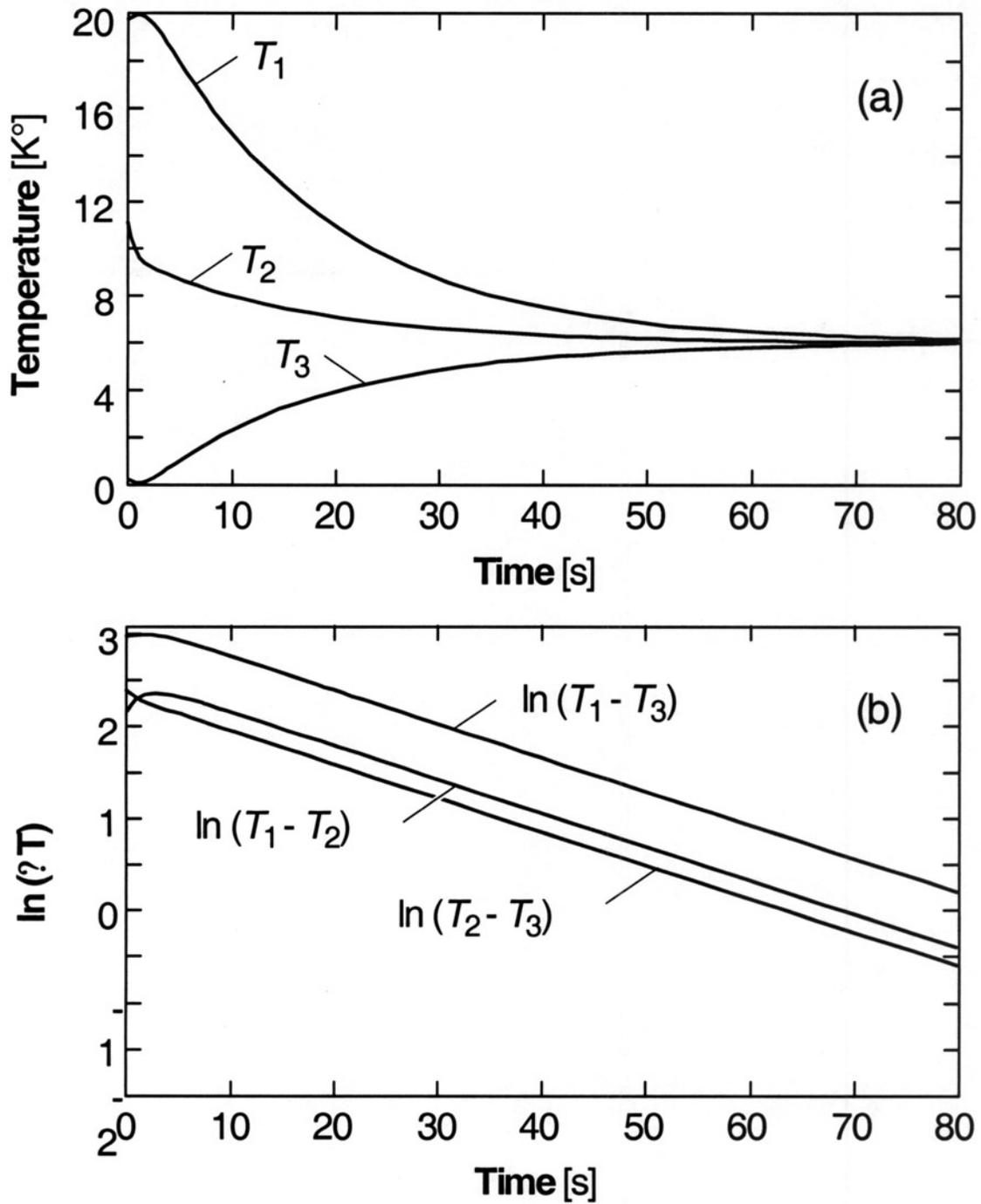


Fig. 2 Temperature profiles and values of $\ln(\Delta T_{ij})$ versus time, generated numerically by using the special heated region and measurement locations. The thermal diffusivity was $0.1 \text{ cm}^2/\text{sec}$. T_i is the temperature at special measurement location r_i .

II-B Mathematical Formulation for Outside Edge Heating

In a similar vein to the final equation derived above for central region heating we arrive at a similar equation for edge heating. We again consider a perfectly insulated cylinder under conditions for which the temperature depends only on the radius r . The initial conditions are, initial temperature T_0 throughout the cylinder and, a heat flux Q'' is applied to the surface for a time $0 \leq t \leq t_e$. The heat flux initial condition (at $r=R$) is expressed as:

$$\text{at } r=R: -K \frac{\partial T}{\partial r} = Q'' [1-h(t-t_e)] \quad (9)$$

where the function h is the Unit Step or Heaviside function, and K the thermal conductivity.

The evolution of the temperature as a function of position and time $T(r,t)$ is obtained by solving the transient form of the heat equation in cylindrical coordinates.

$$\frac{1}{r} \frac{\partial T}{\partial r} + \frac{\partial^2 T}{\partial r^2} = \frac{1}{\kappa} \frac{\partial T}{\partial t} \quad (10)$$

The solution is:

$$f = -\frac{Q'' R}{K} \left\{ \frac{2\kappa t}{R^2} + \frac{r^2}{2R^2} - \frac{1}{4} - 2 \sum_{s=1}^{\infty} e^{-\frac{\kappa \alpha_s^2 t}{R^2}} \frac{J_0\left(\frac{r\alpha_s}{R}\right)}{\alpha_s^2 J_0(\alpha_s)} \right\} \quad (11)$$

where α_s are the positive roots of $J_1(\alpha) = 0$. [Caslaw and Jaeger, Conduction of Heat in Solids, 2nd edition, p 329] Taking the result from Carslaw and Jaeger, it is important to note, that their initial condition is $T_0 = 0$. To have a more general case, the initial value T_0 of the temperature is added to this result.

The presence of the Heaviside function in the solution, divides this problem into two cases, one for $t \leq t_e$ during the heating, and one for $t > t_e$ after the pulse is turned off.

We are interesting in the cooling phase only which yields for the temperature distribution:

$$T(r,t) = -\frac{Q'' R}{K} \left\{ \frac{2\kappa t_e}{R^2} - 2 \sum_{s=1}^{\infty} \left[e^{-\frac{\kappa \alpha_s^2 t}{R^2}} \left(1 - e^{-\frac{\kappa \alpha_s^2 t_e}{R^2}} \right) \right] \frac{J_0\left(\frac{r\alpha_s}{R}\right)}{\alpha_s^2 J_0(\alpha_s)} \right\} + T_0 \quad (12)$$

Only the $n=2$ term can be set to zero, by choosing the temperature measurement locations r_i , in this heating geometry. This is done by setting the $r\alpha_2/R = v_n$ terms to the first two zeros of J_0 , or at $J_0(v_n) = 0$ at $v_n = 2.4048$ and 5.5201 . With $\alpha_2 = 7.01559$ the measurement locations are then set at

$$\frac{r_1}{R} = \frac{v_1}{\alpha_2} = 0.342779 \quad \text{and} \quad \frac{r_2}{R} = \frac{v_2}{\alpha_2} = 0.786833$$

Other terms cannot be so eliminated, however, terms for $n \leq 3$ decay rapidly with time in comparison to the first two terms. Grouping the $n \leq 3$ terms together as higher order terms yields

$$T(r,t) = -\frac{Q'' R}{K} \left\{ \frac{2\kappa t_e}{R^2} - 2 \left[e^{-\frac{\kappa \alpha_1^2 t}{R^2}} \left(1 - e^{-\frac{\kappa \alpha_1^2 t_e}{R^2}} \right) \right] \frac{J_0\left(\frac{r\alpha_1}{R}\right)}{\alpha_1^2 J_0(\alpha_1)} \right\} + h.o.t. \quad (13)$$

Taking the temperature difference between two locations, T_i and T_j :

$$T(r_i, t) - T(r_j, t) = -\frac{Q'' R}{K} \left\{ -2 \left[e^{-\frac{\kappa \alpha_1^2 t}{R^2}} \left(1 - e^{-\frac{\kappa \alpha_1^2 t_e}{R^2}} \right) \right] \frac{J_0\left(\frac{r_i \alpha_1}{R}\right) - J_0\left(\frac{r_j \alpha_1}{R}\right)}{\alpha_s^2 J_0(\alpha_1)} \right\} + \Delta h.o.t \quad (14)$$

This reduces to:

$$T(r_i, t) - T(r_j, t) = e^{-\frac{\kappa \alpha_1^2 t}{R^2}} \times \frac{2Q'' R}{K} \left(1 - e^{-\frac{\kappa \alpha_1^2 t_e}{R^2}} \right) \frac{J_0\left(\frac{r_i \alpha_1}{R}\right) - J_0\left(\frac{r_j \alpha_1}{R}\right)}{\alpha_s^2 J_0(\alpha_1)} \quad (15)$$

Then taking the natural logarithm of the difference we arrive at a solution analogous to equation (7) above:

$$\ln(\Delta T_{ij}) = -\frac{\kappa \alpha_1^2}{R^2} t + \ln(\beta_{ij}) \quad (16)$$

Thus, for either heating geometry we arrive at a solution where only the measured temperature difference versus time is required to calculate the thermal diffusivity.

II-C Modification of the Heated Region-Central Region Heating

We investigate the effect of varying the size of the heated region on the output diffusivity. Initially, this work was intended to see how much error could be tolerated in the heated area; recall that our derivation is based on a special value $p = 0.546$, leading to elimination of the $n = 2$ term in Eq. (5). Here again, we show the results for $\kappa = 0.1$ cm²/sec; the results for the other values of κ follow with linear time scaling. The sample radius for these cases was 5 cm, so the special value of p corresponds with a radius of the heated region of 2.73 cm. Two, extreme, cases are shown here; 1) when the heated region is increased to 125% of the special radius, and 2) decreased to 20%. Thus, the radii of the heated regions considered here are 3.41, and 0.556 cm, respectively.

The temperature increase at equilibrium is given by $T_0 p^2$. Therefore, for an initial temperature of 20 K, the final temperature increases for the exact solution and the two modified cases are expected to be 6.0, 9.3 (125%), and 0.2°C (20%), respectively. The effect of these values on the experimental diffusivity was mentioned before and will be further discussed below.

Figs. 3a, and 4a show a plot of the temperature versus time at the three measurement locations for the modified heated radii (125%, and 20% respectively). These can be compared with Fig. 2 which shows the temperature versus time for the heated region of special radius. The effect of changing the size of the heated region on the measured temperature at r_2 is obvious in Figs. 3a, 4a. Note that for the 0.5 cm heated region (20% of the special p value) the initial T_0 is not seen in the plot since there is no measurement location within this area, Fig. 4a. This change follows what would be intuitively expected, i.e., for the larger heated region, $T(r_2, t)$ is higher, while it is lower for the 20% case (i.e., $p = 0.11$ case). **Note that the approximate time to reach equilibrium is not changed.**

The corresponding plots of $\ln(\Delta T)$ versus time are shown in Figs. 3b, and 4b. Comparing Fig. 2a (special p), at the shorter times, with the modified cases shows the effect of the $n = 2$ term being non-zero on the development of the curve. In Fig. 2b the plot of $\ln(\Delta T)$ versus time becomes linear after about $t > 5$ sec. In Fig. 3b, and 4b, it is not until about 10 seconds that the plots become linear and the input diffusivity is

recovered. For the 125 % case, the time period over which the plot is linear is approximately the same as for the special p case and corresponds to the original diffusivity value to within 1%.

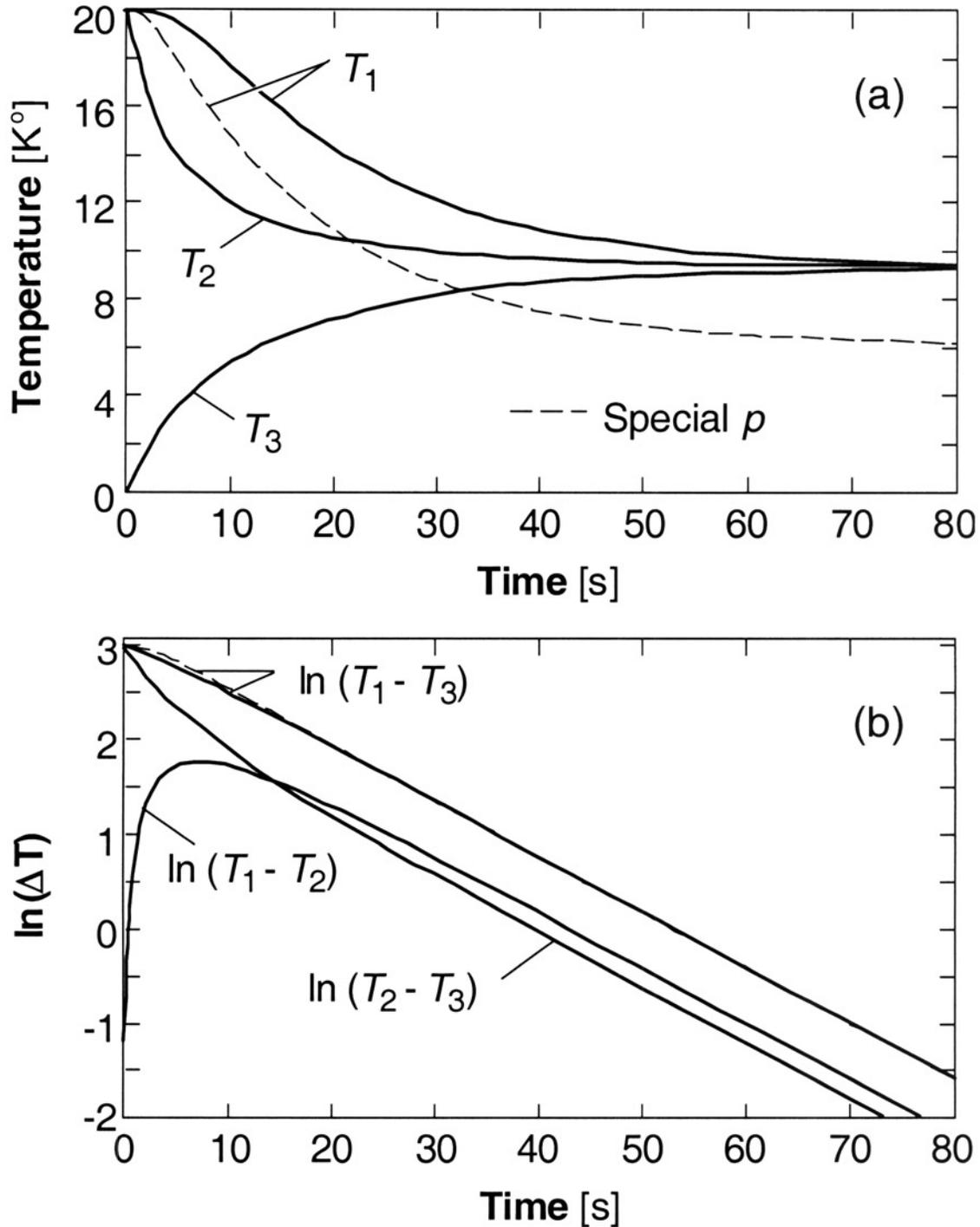


Fig. 3 Temperature profiles and values of $\ln(\Delta T_{ij})$ versus time, generated numerically by using a heated radius 125% larger than the special value and the special measurement locations with no noise. Notice the increase in the temperatures versus time compared to Fig. 2. Figure (b) shows that longer time is required before the diffusivity can be extracted due to the fact that r_2 is no longer at the edge of the boundary between the heated and unheated regions.

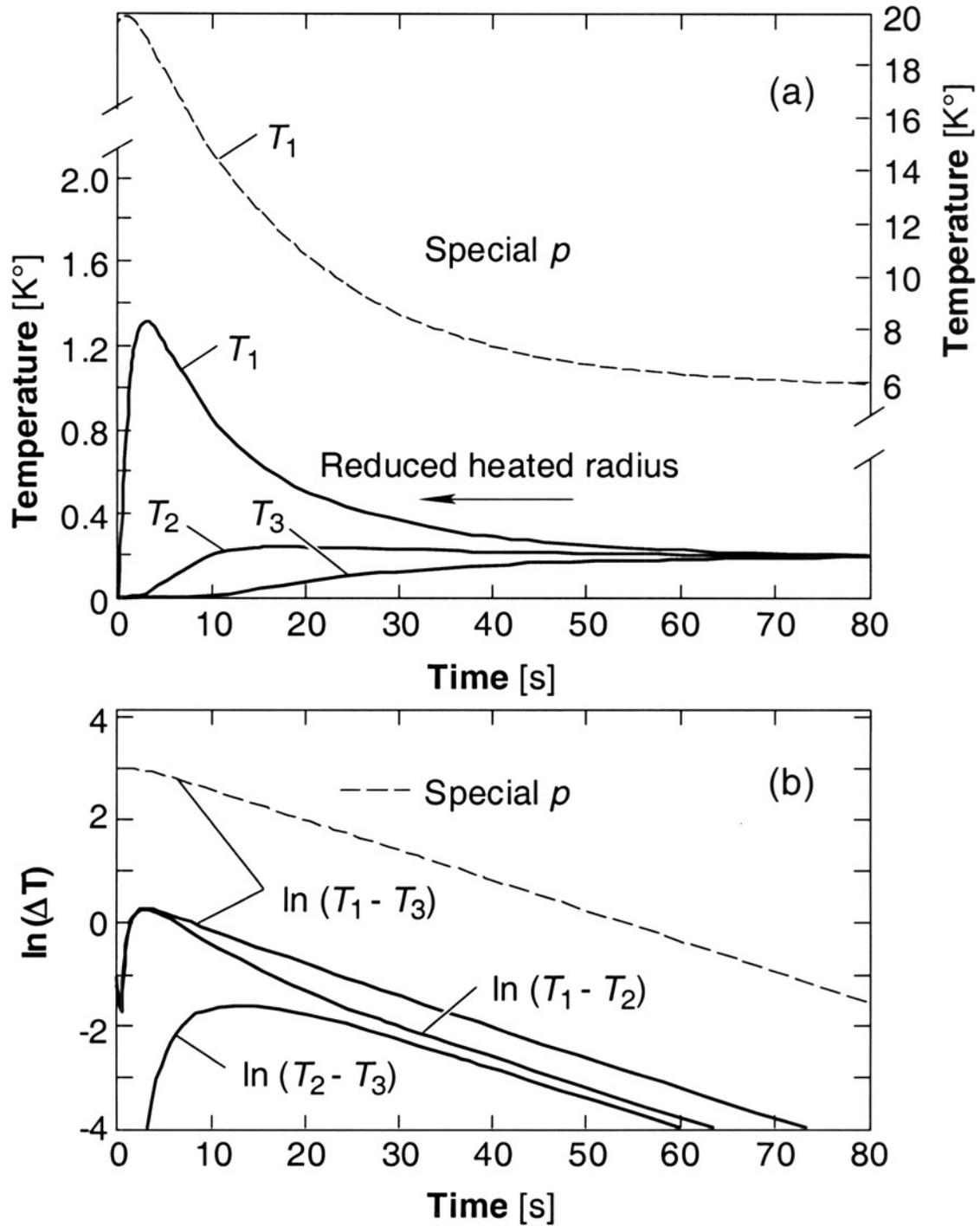


Fig. 4 Temperature profiles and values of $\ln(\Delta T_{ij})$'s versus time generated numerically by using a heated region of size 20% of special value and the special measurement locations with no noise. Note the very small increase in T_1 since the heated region is inside this measurement location. Due to the low values of ΔT the thermal diffusivity can only be extracted from $\ln(\Delta T_{13})$.

II-D Variations in Measurement Locations

The effect of variations in the measurement locations is essentially the same as that for modifications in the heated region, i.e., at long enough times the effect is negligible. This can be seen from Fig. 5a and 5b. Fig. 5 shows the evolution of the temperature profile and $\ln(\Delta T)$ versus time for r_1, r_2 and r_3 of 0.1, 2.5 and, 4.9 cm, (the special r_i values are 1.182, 2.71 and 4.253 cm for $a = 5.0$ cm). As in the above cases, after 10

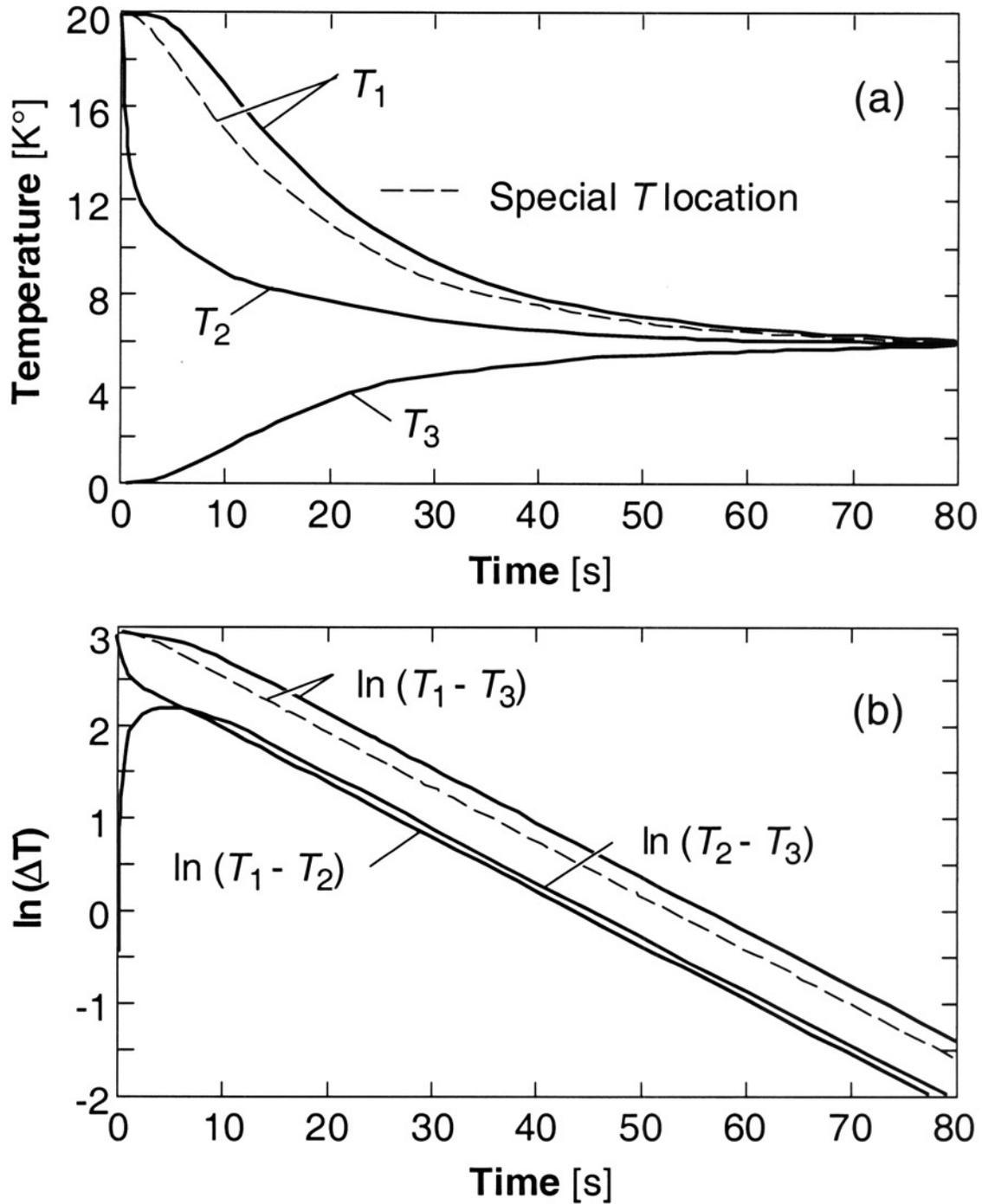


Fig. 5 Temperature profiles and values of $\ln(\Delta T_i)$'s versus time generated numerically by using a heated region of the special size and the modified measurement locations. T_1 and T_3 are inside and outside the correct positions. (b) Longer time is require before the diffusivity can be extracted.

seconds the original diffusivity values are returned. The difference in the temperature profiles at r_1 , r_2 and r_3 between a solution containing 20 terms and a solution containing the constant term $T_0 p^2$ and the $n = 1$ term is less than 0.01 % for values of the non-dimensional coefficient $\kappa t/a^2$ greater or equal than 0.04.

Conclusions

We have developed novel methodology(ies) for measuring thermal diffusivities. This methodology does not require knowing the initial temperature increase or any timing between the applied and measured response. We have shown by numerical simulation that a relative imposed error up to 10% can be tolerated. We have proven that it is possible to extract an accurate value of thermal diffusivity when the heated region is reduced up to 20% of the special value needed to simplify the analytical series solution. Increasing the initial temperature and using modified measurement locations is helpful. Finally, by scaling analysis we have a measure of the time when the thermal diffusivity values are reliable.

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